Unraveling the History of Uranium Contamination to the Vadose Zone in the T-WMA, Hanford Site, Washington

John N. Christensen, P. Evan Dresel[1], R. Jeff Serne[1], Mark Conrad, and Donald J. DePaolo
[1]Pacific Northwest National Laboratory (PNNL)
Contact: John N. Christensen, 510/486-6735, JNChristensen@lbl.gov

RESEARCH OBJECTIVES

For decades, the Hanford, Washington, site was used to process nuclear fuels for the production of plutonium. These past industrial-scale activities resulted in local contamination of the vadose zone and groundwater. An important consideration in the process of cleanup is the source and fate of such contaminants as uranium (U). Our previous work in the B-BX-BY waste management area demonstrated the power of high-precision U isotopic measurements in providing practical constraints on the sources of U contamination, as well as on the behavior and transport of U in the vadose zone. Here we extend this work to two cores that sampled contaminated vadose zone sediments in the T and TX waste management areas (WMAs), laying the groundwork to trace the source of recently recognized ^{99}Tc groundwater contamination in the vicinity of the T-WMA.

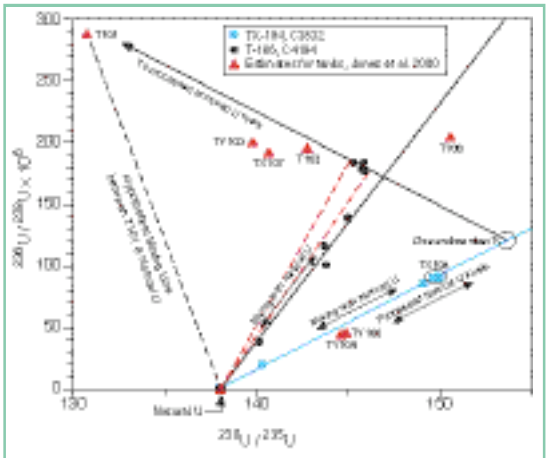

Figure 1. Plot of ^{238}U/^{235}U vs. ^{236}U/^{238}U × 10^6. Black circles are analyses of U from pore water from core C4104 near T-106, blue squares are analyses of pore water from core C3832, near TX-104. Also shown are estimates of tank waste compositions for the T-TX-TY WMAs at their declared leak dates.

APPROACH

The isotopic composition of natural U contrasts with the variable isotopic composition of U from processed fuel rods. This variation can be used as a tracer and fingerprint of contamination. Samples were analyzed for U isotopic composition from two cores (C4104 and C3832) near two single-shelled tanks, T-106 and TX-104, respectively. Uranium from pore waters in the sediment samples were separated and analyzed for isotopic composition by a multiple-collector inductively-coupled-plasma-source mass spectrometer (MC-ICPMS). The results of the isotopic analyses were evaluated and compared to each other to identify sources and the extent of mixing with background U. The isotopic data were also compared to models of the U fuel processing history to provide some temporal constraints.

ACCOMPLISHMENTS

The U isotopic analyses for Core C4104 near Tank T-106, known to have leaked over a six-week period in 1973, indicate that the contaminant U introduced to the vadose zone at this location had a component of processed enriched U fuel, along with a component of processed natural U fuel (Figure 1), at most about 20% and at least 80%, respectively. Moreover, consideration of ^{234}U/^{238}U data (not shown here) constrains the composition of the processed natural U fuel end member. The proportion of processed enriched U decreases down core, until the U isotopic compositions fall along a mixing array with background natural U. In the case of the U isotopic analyses for Core C3832 near Tank TX-104, most of the data form a tight cluster on the array, representing natural U fuels.

SIGNIFICANCE OF FINDINGS

For vadose zone U contamination associated with tanks BX-102 (BX WMA, 200 East Area) and TX-104 (TX WMA, 200 West Area), we found that the contaminant U isotopic composition was highly homogeneous. In contrast, the core near T-106 shows a more complex history, involving mixtures of natural U fuel and enriched fuel, with variable proportions apparently through time. It appears either that contamination was decanted from a tank stratified in U isotopic composition, or that the vadose zone contamination represents mixing with near-contemporaneous contamination from nearby Tank T-103. In either case, the down-core mixing array with background U suggests that the earlier contamination had a lower proportion of processed enriched fuel, providing a basis for modeling the mobility of U in the vadose zone. The data for C4104 also establishes the U isotopic signatures of the waste leak associated with T-106, providing an important part of the isotopic context needed to evaluate the source of ^{99}Tc groundwater contamination in the vicinity of the T-WMA.

RELATED PUBLICATION

Christensen, J.N., P.E. Dresel, M.E. Conrad, K. Maher, and D.J. DePaolo, Identifying the sources of subsurface contamination at the Hanford Site in Washington, using high-precision uranium isotopic measurements. Environ. Sci. Technol., 38 (12), 3330–3337, 2004. Berkeley Lab Report LBNL-54979.

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary of the Office of Environmental Management, Office of Science and Technology, Environmental Management Science Program, of the U.S. Department of Energy, under Contract No. DE-AC03-76SF00098 to LBNL and Contract No. DE-AC06-76RL01830 to PNNL.